Minimizing CO_2 emissions with renewable energy: A comparative study of emerging technologies in the steel industry

Marian Flores-Granobles[a] and Mark Saeys*[a]

CO_2 emissions from the steel industry are amongst the most difficult to abate, since carbon is used as a stoichiometric reducing agent in most steel mills. This carbon ends up as a CO/CO_2 mixture in the steel mill gases, which are combusted to generate heat, electricity, and more CO_2. Strategies to capture and store (CCS), utilize (CCU) or avoid CO_2 in steel production exist, but are highly dependent on the availability of renewable electricity for the production of low-carbon H_2. Steel mill gas contains energy, and can thus be re-used more easily than combustion gas or process gas from the cement industry. In this study, we evaluate several strategies to reduce CO_2 emissions in the steel industry and rank them according to their renewable electricity requirement. We propose the following steps: 1. Shut down the steel plant's power plant, since it produces electricity with a carbon intensity that is even higher than coal-based power plants; 2. Replace steel mill gas with natural gas to generate heat within the steel mill; 3. Recover the reducing gases, H_2 and CO, from the steel mill gases: e.g., using Pressure Swing Adsorption to obtain a H_2-rich stream from COG, and Sorption-Enhanced Water Gas Shift to obtain a H_2-rich stream and a pure CO_2 stream from BFG and BOFG; 4. The recovered H_2 converts some of the CO_2 to methanol, excess CO_2 is stored. The proposed CCUS scenario can retrofit existing infrastructure, uses proven technology and reduces CO_2 emissions by 70% for a marginal renewable electricity demand. Other energy-intensive alternatives have the potential to reduce CO_2 emissions by 85%, but require an order-of-magnitude more renewable electricity.

Broader context

Stoichiometrically, carbon-based steel making would produce 0.6 t CO_2/t liquid steel to reduce hematite to iron. Thermodynamic equilibria between C, CO, and CO_2 and the heat demand of the various processes increase these emissions to 1.9 t CO_2/t liquid steel for a reference steel mill. European regulations drive steel-makers to drastically reduce CO_2 emissions, while staying competitive in a global market. Decarbonising options include capture and storage (CCS), capture and utilization (CCU) and the replacement of carbon by hydrogen as the reducing agent. These options rely on the availability of large amounts of electricity. In this study, we evaluate the CO_2 reduction potential and the electricity requirement for 8 scenarios. We show that, already for a mixed grid, CCUS scenarios can reduce emissions by 55%, and this for a modest electricity demand. By comparing several scenarios, this study provides decision-makers with a road map to reduce CO_2 emissions, while considering the availability and the carbon intensity of electricity.

Introduction

As the dominant material for the construction and automotive sector, steel is critical for economic development[1]. It is strong, resistant, and can be completely recycled[2]. The key step of steel-making occurs in the blast furnace, where carbon removes oxygen from the iron ore. This carbon leaves the process mostly in the Blast Furnace Gas (BFG) which consists of N_2 and equal amounts of CO and CO_2. Steelmakers take advantage of the residual energy in the steel mill gases (in the form of CO) by burning them to generate heat and electricity within the plant. Together, the steel industry is responsible for approximately 7% of the global CO_2 emissions, and is one of the largest industrial point sources[3]. Because of the inherent chemistry of iron ore reduction with carbon, the industry would require complete reconstruction to avoid CO_2 emissions[4].

While steel can be completely recycled, economic development continues to increase global demand for steel, and steel production is expected to grow by 30% over the next 30 years[5]. Decarbonizing technologies must therefore be implemented to reduce CO_2 emissions from steel-making.

Stoichiometrically, 0.6 t CO_2 per t of liquid steel (t l.s.) would be produced in the Blast Furnace to reduce iron ore with coke to hot metal and CO_2. However, the redox equilibrium between coke, CO and CO_2 (i.e., the Boudouard reaction) favours CO above 700 ^oC, and prevents the full conversion of coke to CO_2[6]. In addition, coke is combusted to generate heat in the Blast Furnace[7]. The top gas from the Blast Furnace hence typically consists of 45% N_2, 23% CO, 23% CO_2, 5% H_2 and some water, which makes it a gas with low calorific value. If all this BFG is combusted, steel production would emit around 1.3 t CO_2/t l.s.[8].

Several process modifications have been proposed to reduce CO_2 emissions from the steel industry. CO_2 can be captured from the power plant of the steel mill or from the steel mill gases for its utilization in CO_2-consuming processes (CCU), for storage (CCS) or both (CCUS). H_2-based technologies are emerging due to the increasing availability of renewable electricity, and the possibility to produce carbon-free H_2 by water electrolysis. H_2 could replace carbon as the reducing agent in H_2-based steel-making or it could be used to produce

[a] *Laboratory for Chemical Technology, Ghent University, Technologiepark 125, B-9052 Ghent, Belgium.*

Electronic Supplementary Information (ESI) available: Detailed information about input data for mass and energy balances of the scenarios.

chemicals such as methanol from the captured CO_2 in CCU scenarios. CCS scenarios are somewhat less dependent on electricity to capture CO_2 and transport it to a storage facility, but they do not produce value-added chemicals.

The availability of renewable electricity is hence key to reducing carbon emissions[9, 10], and strategies that require the production of H_2 by electrolysis rely completely on it. However, considering the limited availability of renewable electricity in the foreseeable future and the significantly larger global CO_2 reduction that can be achieved if the available electricity is prioritized for heat generation or mobility instead of electrolysis[11], one may wonder whether massive amounts of renewable electricity should be used for industrial steel production. In this study, we evaluate strategies to reduce CO_2 emissions from steel production that minimize renewable electricity demand, so that it can be used where it most effectively combats climate change.

Current Scenario

A reference integrated steel mill was selected from a study by the International Energy Agency (IEA)[8]. This base case is termed 1-CST (Conventional STeel mill) and the process diagram is shown in Figure S1. The main sections of the reference steel mill are the coking plant, the sinter plant, the pellet plant, the blast furnace, the basic oxygen furnace, the lime plant, the air separation unit and the power plant. The power plant has a thermal efficiency of only 32%[8]. The CO_2 emissions from the ladle metallurgy, the continuous casting and the rolling sections were not considered, nor the emissions corresponding to the extraction and transport of iron ore and coal.

Raw materials are mixed in the blast furnace where liquid hot metal with a carbon content of approximately 4% is produced. Liquid steel is produced in the Basic Oxygen Furnace (BOF) when pure oxygen from an air separation unit is injected in the liquid hot metal from the Blast Furnace to reduce the carbon content to below 1%. The steel mill produces three main gases: Coke Oven Gas (COG), Blast Furnace Gas (BFG) and Basic Oxygen Furnace Gas (BOFG). BFG has the largest volume and the lowest heating value,

Table ***1***. Since steel mill gases contain CO, they cannot be released to the atmosphere. To recover some of the energy from these gases while converting the toxic CO to CO_2, steel mill gases are burned to generate heat and electricity for use in the steel mill.

In the reference steel mill, approximately 1.9 t CO_2 is emitted per t liquid steel[8]. 50% of these emissions are released by the power plant, 30% by the generation of heat in the steel plant, and 20% by other sections of the steel plant such as the sinter, pellet and lime plants. 470 kWh/t l.s. of electricity is produced in the power plant, half of which is consumed in the steel plant, the rest is exported. No CO_2 credit is given for the production of the exported electricity since its carbon intensity is higher than for electricity from the grid.

Table 1. Composition, volume and lower heating value of the steel mill gases[8]

Wet basis (%vol)	*COG*	*BFG*	*BOFG*
CH_4	23.04	-	-
H_2	59.53	3.63	2.64
CO	3.84	22.34	56.92
CO_2	0.96	22.10	14.44
N_2	5.76	48.77	13.83
O_2	0.19	-	-
H_2O	3.98	3.15	12.16
Other HC	2.69	-	-
Volume [Nm^3/t l.s.]	166	1467	82
Lower Heating Value (wet) [MJ/Nm^3]	17.33	3.21	7.47

Opportunities and Challenges

The electricity produced in the power plant of the steel mill has a carbon intensity of around 2.1 t CO_2/MWh[8]. This is much higher than the EU average (0.34 t CO_2/MWh produced[12]) or the carbon intensity of pulverized coal plants (0.8 t CO_2/MWh produced[13]). Shutting down the steel mill power plant and using electricity from the grid (0.42 t CO_2/MWh consumed in EU average grid[12]) would hence reduce CO_2 emissions from steel production by 22%, because CO, CH_4 and C_xH_y in the steel mill gases are no longer combusted and emitted as CO_2.

CO_2 emissions from steel production can be reduced by an additional 7% if natural gas instead of steel mill gas is used to produce heat in the plant. Electrical heating could allow a further reduction[14], but this technology remains to be developed at scale. To achieve these reductions, however, alternative processes need to be implemented to handle the steel mill gases and utilize or store the carbon in these gases.

Instead of burning, CO and H_2 in the steel mill gases could replace fossil raw materials, supporting a cleaner route[15] to chemicals through industrial symbiosis[3, 16, 17]. The integration of CO_2-emitting and CO_2-consuming industries, e.g., the steel and the methanol industry, has been proposed to reduce CO_2 emissions[18]. To tackle the intermittency of renewable electricity, methanol could also be used to chemically store excess renewable electricity[18, 19]. Methanol is one of largest-scale base chemicals[20, 21] and has the highest carbon fuel exergy content per mol of carbon among CO_2-derived liquid fuels[22]. By 2021, the global annual methanol demand is estimated to reach 95 Mt[23]. Methanol production via CO hydrogenation is a mature technology[24], but methanol can also be produced by CO_2 hydrogenation[17]. Since CO_2 is more stable than CO, more energy is required to produce methanol from CO_2[17]. Another option for the valorisation of steel mill gases to chemicals is ethanol. Recently, Lanzatech™ and a leading Chinese steel producer united efforts for the synthesis of ethanol through fermentation of steel mill gases in a commercial plant of 46 kt/year[25]. However, methanol was the selected chemical for this study.

Since CO_2 is not utilized at the pace that it is produced[10], CO_2 storage has to be considered alongside CO_2 utilization in many decarbonization strategies[19]. For storage, CO_2 is compressed to its supercritical state (above 73.8 bar[10]) and transported as a liquid by pipe to the storage site[26]. In CCUS scenarios, the production of methanol in the steel plant is envisioned to provide additional income, which partially offset the cost of CO_2 storage[27].

Commercial installations for CO_2 storage are located mostly in the USA and currently have a capacity of around 30 Mt CO_2 per year[20, 28]. They are mainly developed for Enhanced Oil Recovery. In Europe, the capacity is less than 5 Mt CO_2 per year[18, 28]. The global potential for CO_2 storage is however large, with estimated storage capacities of 3500 Gt CO_2 or more[28, 29].

Treatment of steel mill gases

A variety of separation technologies can recover CO, H_2 and/or CO_2 from the steel mill gases for their subsequent treatment and use. For instance, CO could be recovered by the COSORB^{SM} absorption[30] or CO-PSA[31] but only the emissions for its combustion can be avoided if no additional separation techniques are implemented. CO_2 can be captured by amine scrubbing[32] and either be stored (CCS) or utilized (CCU) for the synthesis of chemicals. CCU requires energy in the form of reductants (e.g. CH_4 as in Super-Dry reforming[33], H_2 as in CO_2 hydrogenation[18]) or electricity as in CO_2 electrolysis[34], which makes it an energy intensive route. Because of the variety of technologies available and broad range of possible process configurations, this study aims to evaluate fairly simple scenarios with mature technologies that offer a large potential reduction of the CO_2 emissions in the steel industry.

Amine scrubbing. A mature and viable technology to capture CO_2 from flue gases is amine scrubbing[17]. The high CO_2 concentration in BFG allows CO_2 capture at a reasonable cost[32]. CO_2 scrubbing from BFG has been demonstrated in the DMX™ CO_2 capture process using amine-based solvents[32]. When BFG is compressed to 6 bar, amine scrubbing requires about 2.4 GJ/t $CO_{2\,captured}$ to regenerate the amines[32]. Amine scrubbing and CO_2 storage are an end-of-pipe decarbonization option and can be implemented by retro-fitting existing steel plants.

Pressure Swing Adsorption (PSA). For more than 50 years[35], high purity H_2 has been produced by PSA[36], and various process configurations can be found in the patent literature. A recent patent claims a H_2 recovery of 90% from gases with more than 50% of H_2 content[37], such as COG.

In this study, a PSA unit was modelled using COG at 35 bar and with a H_2 recovery of 90%, based on a recent patent[37]. The off-gas of this process is a CH_4-rich stream that could be used as a feedstock to produce additional H_2 by steam reforming[38]. In this study, the CH_4-rich stream is exported.

Sorption-Enhanced Water Gas Shift (SEWGS). BFG and BOFG have a high CO and CO_2 content. In the SEWGS process, the residual CO is converted to H_2 via the water gas shift reaction, while simultaneously separating the CO_2[39]. SEWGS is hence a separation process which combines the water gas shift reaction with pressure swing adsorption over a solid CO_2 sorbent[40]. The process comprises a pre-shift section with a WGS catalyst and a sorption column with a potassium-promoted hydrotalcite-based material which acts both as a sorbent and a WGS catalyst[39, 41]. In the pre-shift section, CO reacts with steam and is partially converted to H_2 and CO_2. The sorption column shifts the WGS reaction to full CO conversion by the removal of CO_2. SEWGS has carbon capture ratios above 90% and the produced CO_2 stream is ready for storage, with purities above 90%. SEWGS is at a Technology Readiness Level (TRL) of 6, and has been demonstrated in a pilot plant using BFG from a nearby steel plant[42].

Membrane separation of H_2/N_2 mixtures. The H_2-rich stream from the SEWGS process contains approximately 35% H_2, diluted with N_2 and some CO and CO_2 impurities. To upgrade this stream for use in a methanol synthesis loop, H_2/N_2 separation is required. PSA is not an economical option for gases containing less than 50% H_2[35], therefore membrane separation was considered[43]. A two-stage purification process was modelled using relative permeabilities from literature[44].

Renewable Energy

The introduction of renewable energy in industrial processes is an important route to reduce carbon emissions[9, 10, 45]. This route clearly requires a dramatic expansion of the infrastructure to collect renewables such as wind, solar and hydro power. Even then, renewable electricity is not expected to fully cover electricity demand in the foreseeable future. The introduction of solar and wind energy has however steadily reduced the grid emissions intensity (GEI) in the European Union to the current level of 0.42 t CO_2/MWh consumed[12]. This is lower than the global GEI of 0.52 t CO_2/MWh produced[46]. Sweden, France, Finland and Belgium have GEIs below the EU average; Sweden emits 0.04 t CO_2/MWh consumed and Belgium 0.26 t CO_2/MWh consumed[12]. In this study, 2 GEIs have been considered: GEI_1, 0.25 t CO_2/MWh, is typical for a mixed grid with fossil and renewable energy, and comparable to the one in Belgium at the moment of this study, GEI_2, 0.01 t CO_2/MWh, is representative for a future grid with only renewable energy[47], and is projected to be available only after 2060[46].

H_2 production by electrolysis. Water electrolysis offers a route to low-carbon H_2. A significant barrier to the introduction of large-scale electrolysis units is the availability of abundant, low-cost renewable electricity[38, 48]. In our model, 4.5 kWh of electricity is required to produce 1 Nm^3 of H_2[38].

H_2-based steel-making. The reduction of the iron ores by H_2 could largely avoid CO_2 emissions by steel plants. In H_2-based steel-making, equipment for the direct reduction of iron ore is required: a direct reduction reactor and an Electric Arc Furnace (EAF) replace the Blast Furnace and Basic Oxygen Furnace. Iron oxide pellets are fed to the direct reduction reactor where H_2 transforms the pellets into a 'sponge iron'. In contrast with the Blast Furnace route, the metal is not melted during the direct reduction. This solid 'sponge iron' is then fed to an EAF to produce liquid steel. CO_2 emissions per t l.s. can be considerably lower in H_2-based steel-making when renewable electricity is used in the EAF. The production of pellets also emits less CO_2 than the production of sinter[49].

The use of H_2 as the reductant in steel-making is not yet a mature technology[17] and requires further improvements in particular for the dynamic control of the process and the prevention of the stickiness of the Direct Reduced Iron[50]. Nevertheless, the leading technologies in direct reduction of iron with reducing gases (MIDREX™ [51] and ENERGIRON™) are making efforts to operate with pure H_2 as reducing gas. To date,

the ENERGIRON™ technology has demonstrated the use of H_2-rich gases such as the COG and reformed gas with H_2/CO ratios as high as 5 to reduce iron ore in industrial direct reduction reactors and has tested higher ratios on a pilot scale[49,52]. Since the reduction of iron ore with H_2 is an endothermic reaction, approximately 800 Nm^3 H_2/t l.s. is required to provide both heat and reducing power in a direct reduction plant[53]. In addition to the construction of large scale electrolysers (about 3.6 MWh/t l.s.), completely new infrastructure would need to replace the blast furnaces. Therefore, H_2-based steel-making is not a short-term decarbonization option[50].

In this study, the CO_2 emissions for the production of 1 t l.s. through the H_2-based steel-making include the production of the pellets[49], the production of lime[8], the injection of coal in the EAF[52], the decomposition of the electrodes[54] and the carbon intensity of the consumed electricity. The total electricity consumption for a H_2-based steel plant is 4.2 MWh/t l.s., based on the requirement of 8.7 GJ H_2/t l.s. (with 94% of metallization)[52, 53] and the consumption of 630 kWh/t l.s. in the direct reduction reactor and in the EAF.[53]

Alternative methanol synthesis. Conventional methanol synthesis includes steam methane reforming, CO hydrogenation and distillation. Per ton of methanol, this process consumes 39 GJ of natural gas, requires around 170 kWh of electricity, exports 2 GJ of steam and emits 0.52 t CO_2[47]. An alternative route is via CO_2 hydrogenation using renewable hydrogen. Stoichiometrically, 1.4 t CO_2 is converted to 1 t methanol. This process consumes 1.5 MWh of electricity per t methanol for the compressors and distillation[47].

The Scenarios

Based on the possible decarbonization processes discussed above, 8 scenarios have been developed to evaluate the CO_2 reduction potential and the electricity requirement. A brief description of the scenarios is given in Table 2. For each scenario, the process diagram is shown in Figures S1-S8. The analysis is based on material and energy balances, using data reported in literature and summarized in Tables S2-S6. The literature data are supported by detailed Aspen Plus (V11.0 Aspen Technology) and Matlab (Research R2017b Mathworks) simulations for the various process steps. Aspen Plus simulations used the Peng-Robinson equation of state with modified Huron-Vidal mixing rules.

Alternative steel-making. A steel plant based on the reduction of iron ore pellets with H_2 is evaluated in scenario 2-HST.

CCS. Two scenarios consider the capture and storage of CO_2 from the BFG using amine scrubbers installed at the outlet of the blast furnace, before the power plant. In the first scenario, waste heat available in steel mill is used to regenerate the amines. Steel plants have 0.32 GJ/t l.s. of utilizable waste heat within the boundaries set in this study[55]. However, only 0.05 GJ/t l.s. from the hot blast stoves can be used to regenerate the amines, considering the required temperatures. In scenario 3-CST-WHC only this limited amount of waste heat is used to regenerate the amines, while in scenario 4-CST-EBC both the waste heat and steam produced by an electric boiler with 97% efficiency[47] are used to capture 90% of the CO_2 in the BFG. These scenarios include the compression of BFG to 6 bar[32] and the compression of CO_2 to 110 bar for storage[8].

GPP and GPH scenarios. Steel mill gases are currently combusted in the power and in the steel plant to produce electricity and heat, respectively. Two sets of scenarios were considered: GPP scenarios treat only the Gases used in the Power Plant, while GPH scenarios treat both the Gases used in the Power Plant and the gases used in the steel plant for Heating. The grid now supplies the electricity required in the steel plant. Natural gas with a Lower Heating Value (LHV) of 40.6 MJ/Nm^3 [8] is combusted to generate heat in the GPH scenarios. CO_2 emissions due to the extraction of natural gas were not considered.

CCUS. In the CCUS scenarios, COG is compressed to 35 bar to recover 90% of its H_2 by PSA. The CH_4-rich stream is exported, and the emissions associated with its combustion are avoided in the steel plant. Both the BFG and BOFG are compressed to 26 bar for the SEWGS process and the required steam is supplied by electrical boilers with 97% of efficiency[47]. 90% of the H_2 produced in SEWGS is recovered by a 2-stage membrane separation. The H_2 from the COG-PSA and from the SEWGS process is used to produce methanol via CO_2 hydrogenation.

A ton of methanol produced by this alternative route utilizes 1.4 t CO_2 and requires 1.5 MWh of electricity to drive the compressors and the separation units[47]. Since in these scenarios, the production of methanol via CO_2 hydrogenation replaces methanol produced via the fossil route, a reduction in the CO_2 emissions of 0.52 t CO_2/t methanol[47] is considered, i.e., the direct CO_2 emissions from natural gas-based methanol production. The H_2 streams from the PSA and the membrane separation are used to produce 0.14 t methanol/t l.s. in scenario

Table 2. Overview of the scenarios

Scenarios	Description
1 **CST**	Conventional steel mill with power plant.
2 **HST**	H_2-based steel mill.
3 **CST-WHC**	Conventional steel mill with power plant and pre-combustion CO_2 capture using the steel plant's waste heat.
4 **CST-EBC**	Conventional steel mill with power plant and pre-combustion CO_2 capture using the steel plant's waste heat and steam from electric boilers.
5 **CST-GPP-CCUS**	Conventional steel mill with treatment of the gases originally sent to the power plant, CO_2 storage and methanol production.
6 **CST-GPH-CCUS**	Conventional steel mill with treatment of both the gases originally sent to the power plant and the gases burned for heating, CO_2 storage and methanol production.
7 **CST-GPP-CCU**	Conventional steel mill with treatment of the gases originally sent to the power plant, H_2 production by electrolysis and methanol production.
8 **CST-GPH-CCU**	Conventional steel mill with treatment of both the gases originally sent to the power plant and the gases burned for heating, H_2 production by electrolysis and methanol production.

5-CST-GPP-CCUS and 0.22 t methanol/t l.s. in scenario 6-CST-GPH-CCUS, as shown in Table S1. Because all the steel mill gases are treated in scenario 6-CST-GPH-CCUS, methanol production is higher than in scenario 5-CST-GPP-CCUS, where only the gases initially sent to the power plant are treated. The excess CO_2 from the SEWGS process, i.e., 0.63 t CO_2/t l.s. for scenario 5 and 1.0 t CO_2/t l.s. for scenario 6, are compressed to 110 bar, and sent for storage.

CCU. In the CCU scenarios 7 and 8, additional H_2 is produced by water electrolysis to convert all the CO_2 to methanol. No CO_2 is stored in the CCU scenarios. In scenario 7-CST-GPP-CCU, 0.59 t methanol/t l.s. is produced, while in scenario 8-CST-GPH-CCU, 0.96 t methanol/t l.s. is produced. Additional electricity is required for water electrolysis.

Analysis and Discussion

CO_2 emissions are shown as a function of electricity consumption for each of the 8 scenarios in Fig 1. Fig 1a shows data for a mixed grid while Fig 1b shows data for a fully renewable electricity grid. Figure 2 gives a detailed breakdown of the emitted CO_2 and the stored and avoided CO_2 for each scenario. Detailed material and energy balances are given in Tables S7-S9.

Mixed grid (GEI_1 = 0.25 t CO_2/MWh)

A simple route to reduce carbon emissions from the steel plant is the capture and storage of CO_2 from pre-combustion BFG (scenarios 3 and 4). When only waste heat is used for the regeneration of the amines (scenario 3), the CO_2 emissions reduce by only 0.020 t CO_2/t l.s., equivalent to 1% of the CO_2 emissions from the steel plant. A larger 15% reduction can be achieved when additional steam is produced to regenerate the amines, and this for a limited electricity demand for the electric boilers. The excess electricity produced by the power plant covers the electricity requirement to run the CO_2 capture installation in scenario 3 (but can now no longer be exported to the grid). In scenario 4, 0.14 MWh/t l.s. electricity is required from the grid to operate the CO_2 capture plant, leading to 0.036 t of CO_2 emissions. These scenarios have a high CO_2 abatement per MWh of electricity (Table S10), but their CO_2 reduction potential is limited since the CO in the BFG is still converted to CO_2 in the power plant and emitted. An alternative option is post-combustion capture of the CO_2, but the low thermal efficiency of the power plant, 32%[8], makes this option less interesting.

CCUS scenarios 5 and 6 allow a much larger reduction of the CO_2 emissions, see Fig 2a. In scenario 5, the power plant is shut down, H_2 is recovered from COG in a PSA unit, pure H_2 and CO_2 streams are obtained from BFG and BOFG in a SEWGS process with membrane separation, and the recovered H_2 (280 Nm^3 H_2/t l.s.) is used to convert 0.19 t CO_2/t l.s. to 0.14 t methanol/t l.s. The excess 0.63 t CO_2/t l.s. is ready for storage. In scenario 5, CO_2 emissions decrease by 39% from 1.9 t CO_2/t l.s. to 1.2 t CO_2/t l.s. In scenario 6, natural gas combustion replaces COG and BFG combustion to provide heat in the plant, and additional COG and BFG are sent for treatment. This supplies an additional 180 Nm^3 H_2/t l.s. (and 0.08 t methanol/t l.s.) and an additional 0.39 t CO_2/t l.s. for storage.

Half of the electrical demand of the CCUS scenarios arises from the treatment of the steel mill gases, in particular the gas compressors for the PSA unit, for CO_2 storage, for the membrane separation and for the SEWGS process, along with the electric boilers in the SEWGS process. The other half is required for the methanol plant and the operation of the steel mill plant, since the power plant is shut down in the CCUS scenarios. The total electrical demand is 0.79 MWh/t l.s. in scenario 5 and 1.1 MWh/t l.s. in scenario 6.

A clear advantage of the CCUS scenarios is that the energy contained in the steel mill gases (in the form of H_2 and CO) is used to convert part of the captured CO_2 to methanol, replacing natural gas as a feedstock and fuel in conventional methanol production and avoiding the direct emission of 0.52 t CO_2/t methanol.

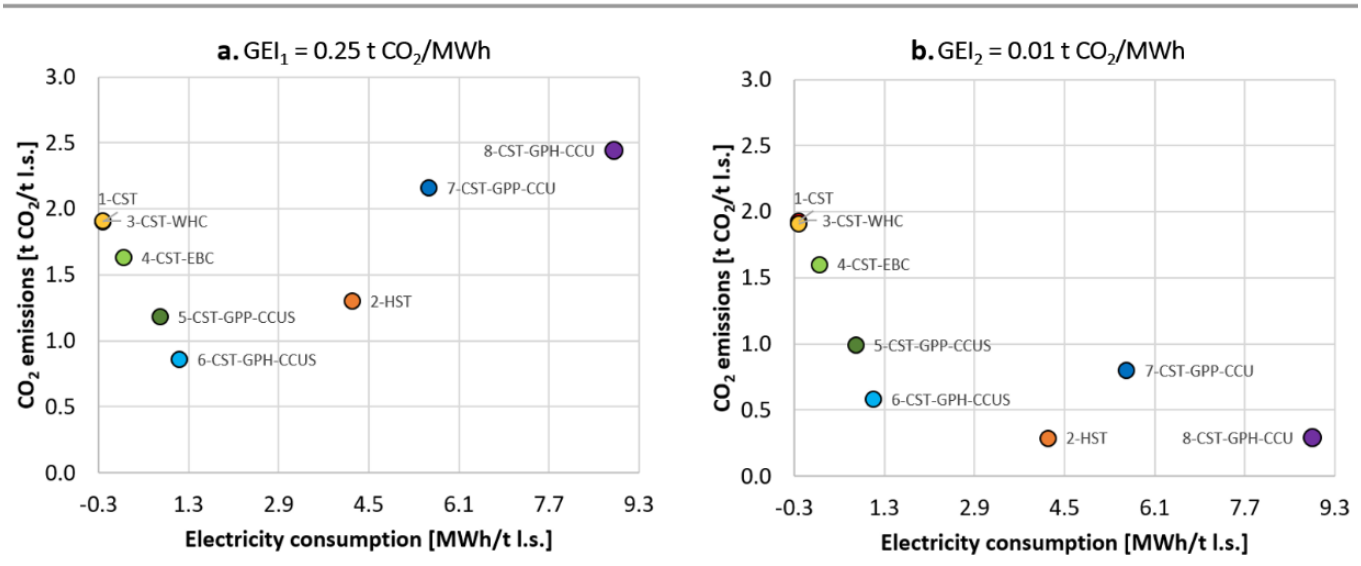

Figure 1. CO_2 emissions and electricity consumption for the 8 scenarios in Table 1, using a mixed grid (a) and a fully renewable grid (b).

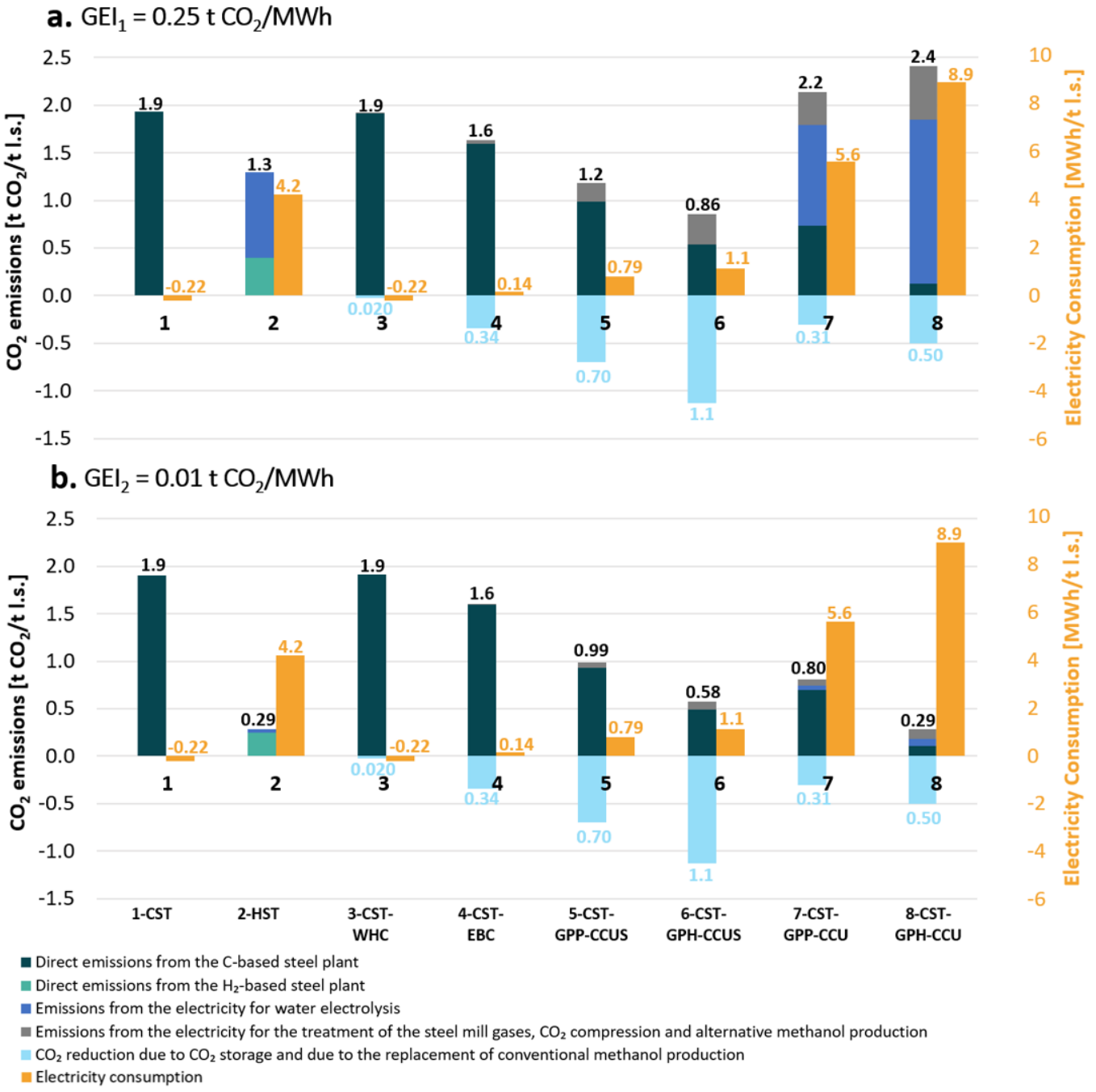

Figure 2. Breakdown of the CO_2 emissions and of the electricity consumption for the 8 scenarios considering a mixed (a) and a renewable (b) grid.

For the mixed grid, the two CCUS scenarios have high efficiencies for the use of electricity for CO_2 abatement. The CCUS strategies reduce the emissions by 39% to 1.2 t CO_2/t l.s. and consume 0.79 MWh/t l.s. electricity from the grid (scenario 5, where only the gases sent to the power plant are treated) or by 55% to 0.86 t CO_2/t l.s. with 1.1 MWh/t l.s. electricity from the grid (scenario 6, where all the gases are treated). Note that these significant reductions can be achieved by retrofitting existing plants, and by using so-called grey electricity from a mixed grid with a carbon intensity of 0.25 t CO_2/MWh. Even larger reductions can be achieved if the fraction renewable electricity increases (Fig S9).

In the CCU scenarios, all CO_2 is converted to methanol with H_2 from water electrolysis. No CO_2 is sent for storage. CCU scenarios are popular in the literature[17, 22, 56] and are investigated in several EU projects[57-59]. Because of their large electricity demand, CCU scenarios do not reduce the CO_2 emissions for a mixed grid (Fig 2a). The CCU scenarios 7 (only gases from the power plant are treated) and 8 (all gases are treated) *increase* emissions to 2.2 t CO_2/t l.s. and 2.4 t CO_2/t l.s., respectively. These scenarios require large amounts of electricity, 5.6 MWh/t l.s. and 8.9 MWh/t l.s., respectively, to produce additional H_2 by electrolysis. About 80% of the electricity is consumed by the electrolysers in these scenarios. Although there is a significant reduction in the *direct* CO_2

emissions from the steel plant to 0.75 t CO_2/t l.s in scenario 7 (dark green bars in Fig 2a) and to 0.17 t CO_2/t l.s. in scenario 8, this is more than compensated by the *indirect* CO_2 emissions associated with electricity production (1.4 and 2.3 t CO_2/t l.s.). The viability of the CCU scenarios hence depends critically on the availability of renewable electricity and only for GEI below 0.19 t CO_2/MWh these scenarios offer a net reduction in the CO_2 emissions (Fig S9).

Hydrogen-based steel-making (scenario 2) requires significant amounts of hydrogen. Even if electricity from a mixed grid feeds the electrolysers, CO_2 emissions in this scenario are 33% lower than for conventional carbon-based steel-making. Total CO_2 emissions are 1.3 t CO_2/t l.s., where 0.24 t CO_2/t l.s. are direct process emissions from the pellet and lime production, the carbon addition to the EAF and the decomposition of the electrodes in the EAF, and 1.1 t CO_2/t l.s. are indirect emissions from electricity consumption in the electrolysers and in the direct reduction process, mostly in the EAF. While H_2-based steel-making reduces CO_2 emissions already for a mixed grid and is more promising than the CCU scenarios, it requires 5 times more electricity to abate a ton of CO_2 than the CCUS routes.

Renewable grid (GEI_2 = 0.01 t CO_2/MWh)

For comparison, we also evaluated the CO_2 reduction potential for a fully renewable grid with a GEI of 0.01 t CO_2/MWh, Fig 1b. This energy intensity is typical for large scale wind farms[47] and is expected to be reached for the EU grid by 2060[46]. The low GEI obviously has the largest impact for scenarios that rely on water electrolysis (CCU and HST). It has no effect on the reference scenario 1 since the electricity is supplied by the steel mill's power plant.

CO_2 emissions in the CCUS scenarios further reduce slightly to 0.99 t CO_2/t l.s. and 0.58 t CO_2/t l.s. (49% and 70% CO_2 emissions reduction, respectively) when the grid intensity decreases to 0.01 t CO_2/MWh, Fig 1b. This reduction results from the lower *indirect* emissions from the electricity used by the various compressors in these processes, Fig 2b. The CCUS scenarios continue to offer the largest reduction in CO_2 emissions per MWh electricity.

While the CCU scenarios 7 and 8 did not reduce emissions for a mixed grid, they reduce emissions by 58% and 85% when renewable electricity is used. The remaining emissions are still substantial at 0.80 t CO_2/t l.s. and 0.29 t CO_2/t l.s., respectively, and come mainly from non-electricity related emissions in the steel plant. The replacement of fossil methanol production by CO_2 hydrogenation accounts for an emission reduction of 0.31 t CO_2/t l.s. in scenario 7 and of 0.50 t CO_2/t l.s. in scenario 8, but fossil methanol production might no longer be economically viable when renewable electricity has become abundant. The CCU scenarios require the most electricity per ton of abated CO_2, as shown in Table S10.

These results are somewhat sensitive to the selected process performance parameters (Table S11), as shown in Figure S15 and S16. For example, 2 key parameters were evaluated for the SEWGS process: the CO_2 recovery and the CO_2 purity. Decreasing the CO_2 recovery from 95% to 90% increases CO_2 emissions for CCUS scenario 6 and for CCU scenario 8 by 11% and 28%, respectively, for a renewable grid. This additional CO_2 is emitted with the N_2-rich stream from the membrane separation after the SEWGS process. Decreasing the CO_2 recovery also reduces the electricity consumption for CCU scenario 8 by 0.5 MWh/t l.s. since less CO_2 needs to be hydrogenated to methanol. Decreasing the CO_2 purity from 95% to 90% has a limited effect on our results but might complicate CO_2 storage. For the PSA unit, decreasing the H_2 recovery from COG from 90% to 70% has a limited effect on the CO_2 emissions of CCUS scenario 6 and CCU scenario 8, but increases the electricity consumption for CCU scenario 8, since more H_2 is required to transform the CO_2, and reduces the electricity consumption of CCUS scenario 6, since the lower H_2 recovery from COG reduces the methanol production slightly by 0.01 t methanol/t l.s. Hydrogen-based steel-making reduces CO_2 emissions as much as the most aggressive CCU scenario 8 for the fully renewable electricity grid, but requires only half the electricity. The reduction is now also larger than what can be achieved in CCUS scenario 6. Hydrogen-based steel-making however depends entirely on the continuous availability of electricity, and it still requires twice as much electricity to abate a ton of CO_2 as CCUS scenario 6.

Economically, the selection of a CO_2 abatement technology will require a careful evaluation of operating and capital costs. The most important operating cost for the different abatement technologies will be the cost of electricity, and hence CCUS scenarios provide an interesting balance between electricity consumption and CO_2 abatement. In addition, the production of methanol generates additional value. H_2-based steel-making provides the same CO_2 emissions reduction as the most aggressive CCU scenario 8, but requires only half the electricity; yet, it does not produce an additional product. In terms of capital expenditure, CCUS and CCU scenarios can be implemented by retrofitting the existing steel infrastructure, while the H_2-based scenario requires substantial capital investments in new infrastructure.

In scenarios 6 (CCUS) and 8 (CCU), the gas treatment plants have the same dimension. One option would be to over-dimension the methanol plant so that CO_2 intended for storage in the CCUS scenario can be converted to additional methanol when H_2 can be produced from surplus renewable electricity. This would offer the steel plant operator flexibility to maximize profits. For example, if surplus renewable electricity is available to produce 500 Nm^3 H_2/t l.s., an additional 0.24 t methanol/t l.s. can be produced. This scenario, intermediate between 6 and 8, reduces CO_2 emissions by 75%, produces 0.46 t methanol/t l.s. and stores 0.68 t CO_2/t l.s. H_2-based steel-making does not offer this flexibility and requires a large and constant supply of electricity to achieve its annual production.

Electrically intensive scenarios 2 and 8 are not recommended without abundant renewable electricity, either from the grid or from dedicated solar or wind farms. A reference steel mill producing 4.4 Mt l.s. per year would require 2.1 GW for hydrogen-based steel-making and 4.4 GW for CCU scenario 8. This corresponds to 5900 3 MW windmills working at 25% of capacity. In CCUS scenario 6, a more reasonable 570 MW is required for a reference steel plant.

Conclusions

Steel-making via the conventional blast furnace route is an energy-intensive process which accounts for 7% of global CO_2 emissions[3]. Drastic and costly CO_2 abatement options are a challenge for a steel industry faced with low profit margins per ton of emitted CO_2. In this study, we evaluate several scenarios to reduce CO_2 emissions from steel-making and rank them according to their electricity needs.

CCS scenarios that remove CO_2 from BFG can only reduce CO_2 emission by 17% and are hence not an option to drastically reduce CO_2 emissions.

The combustion of low caloric steel mill gases to produce power and heat is highly inefficient, with carbon intensities that are six times higher than the EU grid average. In the CCU(S) scenarios, steel mill gases are therefore treated to recover H_2 and CO_2 separately. The recovered hydrogen is used to convert some of the CO_2 to methanol; excess CO_2 is exported for storage or converted to additional methanol using hydrogen produced by electrolysis. Already for a mixed grid with a carbon intensity of 0.25 t CO_2/MWh, the CCUS scenario can reduce CO_2 emissions by 55% for an electricity demand of 1.1 MWh/t l.s. or 570 MW for a 4.4 Mt l.s./year steel plant.

As grid intensities decrease, other scenarios can further reduce CO_2 emissions. For a grid intensity below 0.11 t CO_2/MWh, H_2-based steel-making offers a larger reduction than the CCUS scenario. The maximum reduction of 85% is achieved for a fully renewable grid. Hydrogen-based steel-making however requires completely new installations and hence significant investments.

In the CCU scenarios, no CO_2 is stored, and additional hydrogen is produced by electrolysis to convert CO_2 to methanol. CCU scenarios reduce CO_2 emissions more than CCUS only for grid intensities below 0.05 t CO_2/MWh. They reach a maximum CO_2 reduction of 85% for a fully renewable grid. The electricity demand to abate a ton of CO_2 is very high in CCU scenarios, 5.5 MWh/t CO_2, and five times higher than in CCUS scenarios.

If large amounts of renewable electricity are available for the steel industry, H_2-based steel-making offers the most energy efficient option for the low emission steel plant. However, if this renewable electricity is not available for the steel industry and rather prioritised for heating and mobility, then CCUS scenarios offer the most efficient way to reduce CO_2 emissions. The CCUS scenarios do not require a large share of electricity and can already reduce CO_2 emission by 55% for a mixed grid. They can moreover be implemented by retrofitting existing infrastructure, while H_2-based steel-making requires large investments in new infrastructure. Electricity-intensive H_2-based steel-making and CCU scenarios can therefore only be implemented in the long-term when an extensive and stable renewable grid has become available. CCUS scenarios on the other hand offer the opportunity to reduce CO_2 emissions drastically already in the short-term and for a mixed grid.

Conflicts of interest

There are no conflicts to declare.

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